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X-Ray Free Electron Laser Interaction With Matter

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Abstract. X-ray free electron lasers (XFELs) will enable studying new areas of laser-matter interaction. We summarize the current understanding of the interaction of XFEL pulses with matter and describe some of the simulation approaches that are used to design experiments on future XFEL sources. Modified versions of these models have been successful in guiding and analyzing experiments performed at the extreme-ultraviolet FEL FLASH at wavelengths of 6 nm and longer. For photon energies of several keV, no XFEL-matter interaction experiments have been performed yet but data is anticipated to become available in the near future, which will allow to test our understanding of the interaction physics in this wavelength regime.

Keywords: X-ray free electron laser, x-ray matter interaction
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INTRODUCTION

X-ray free electron lasers (XFELs) hold the promise to produce high-intensity, ultrashort pulses of coherent, monochromatic radiation in the 1 to 10 keV photon energy regime [1-2], enabling many research endeavors. The Linac Coherent Light Source (LCLS) is currently being commissioned and will provide XFEL pulses in the energy range of 1 to 8 keV with a pulse length of less than 200 fs, and a total pulse energy of more than 2 mJ. In many applications, this beam is going to be focused to a diameter of 1 μm or less. These expected light output characteristics will enable us to enter a completely new regime of x-ray matter interaction that has not been tested experimentally so far.

Recently, an extreme-ultraviolet free-electron laser (FLASH) has been built at DESY in Hamburg [3]. This source has allowed us to study the interaction of high-fluence short-duration photon pulses with materials at the shortest wavelength possible to-date, down to 6 nm. With these experiments, we have come closer to the extreme conditions expected in XFEL-matter interaction scenarios than previously possible.

X-RAY MATTER INTERACTION

We will now describe the relevant processes when an XFEL beam irradiates a material. In the x-ray regime around 10 keV, the primary photon-matter interaction mechanism is bound-free absorption (photoionization), primarily of the inner K-shell. Only for larger x-ray energies does inelastic scattering become dominant. In general, materials with a low atomic charge number Z absorb fewer photons than high-Z

materials. We will focus the discussion on materials with low atomic charge number *Z*. Within 5-10 fs, the excited atoms relax through Auger decay, emitting electrons of a few hundred eV energy. The "free" Auger and photoelectrons initially escape the sample. When a sufficiently large positive charge is established, the Auger electrons become electrostatically trapped ("quasi-free"). The photoelectrons are trapped only later in the pulse. The trapped electrons thermalize with each other with in a few fs. The relevant atomic physics processes are shown in Figure 1. Ionization of the material during the XFEL pulse will affect both the photoionization rate and the atomic form factor that determines the elastic scattering from the atoms [4]. The trapped electrons establish a spatial distribution in which the inner region of the molecule is neutralized and the outer layer is highly positively charged. The charged outer layer explodes from the Coulomb force, and a rarefaction wave propagates inward toward the center of the nearly neutral molecules, causing its expansion, see Figure 2.

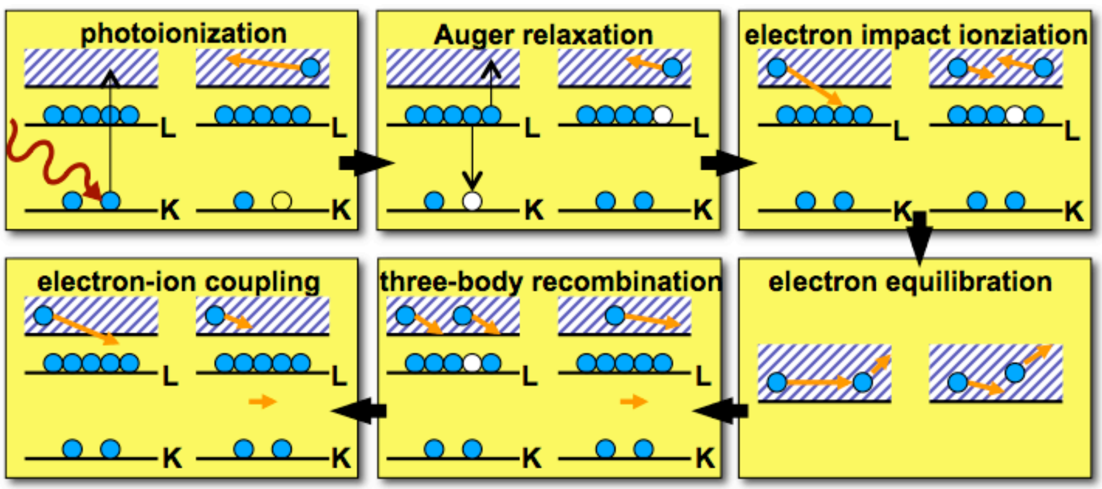

FIGURE 1: Relevant atomic processes in an energy diagram. K and L denote the atomic shells with principal quantum numbers 1 and 2, respectively.

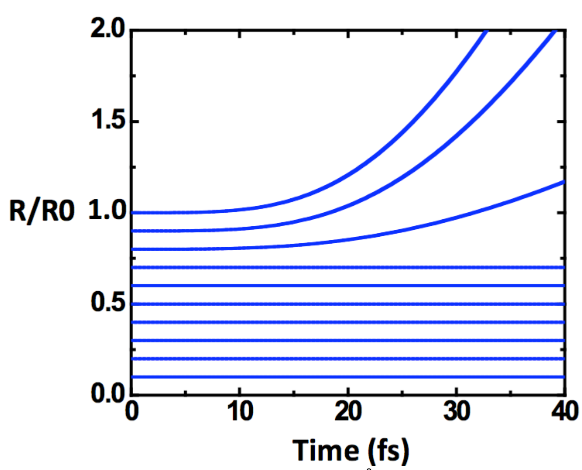

FIGURE 2: Motion of atomic shells for the case of a 60 Å radius molecule illuminated by a photon pulse of flux 1.5 x 10^{11} ph/(100nm)^2/fs for 40 fs.

High-field effects such as multi-photon ionization and excitation, including above-threshold ionization and high-harmonic generation, are well-known phenomena in the optical and, more recently, in the extreme-ultraviolet regime. It is expected that in the x-ray regime, inner-shell phenomena will dominate. Currently, little is known about multi-photon inner-shell x-ray processes.

The linear polarization of the LCLS beam will determine the XFEL-matter interaction volume in macroscopic samples. It is understood that the preferred emission direction of K-shell photoelectrons is in the direction of the incoming electric field. Depending on the angle of incidence, the penetration depth of the x-rays, d_{xray}, the range of the photoelectron, d_{range}, and the lateral straggle of the photoelectron, $d_{straggle}$, determine the excitation volume. The different scenarios are sketched in Figure 3.

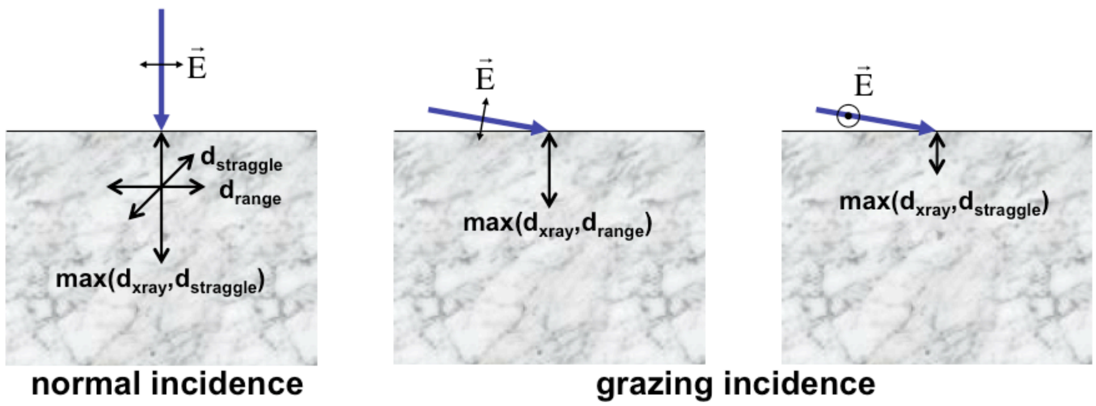

FIGURE 3: Polarization-dependent energy deposition in a semi-infinite solid. The thick arrow indicates the direction of the XFEL beam.

MODELS TO DESCRIBE X-RAY MATTER INTERACTION

A general understanding of the XFEL-matter interaction dynamics has been derived from molecular dynamics [5-7] and continuum dynamics [8-11] models.

Continuum models

Continuum models to describe the dynamics are computationally very efficient and enable the survey of a large parameter space. One specific model described in Ref. [8] is a two-fluid hydrodynamic model that assumes that the sample is initially a homogeneous continuum and that the sample has spherical symmetry. Free electrons and ions are treated as separate fluids that interact by the Coulomb force. Atomic rate equations are used to describe the ionization dynamics of each atomic species. As an improvement to this continuum model, Ziaja et al. has developed a dynamics model based on Boltzmann equations that tracks the velocity distribution [9]. Both continuum models lack the atomic detail of MD.

Molecular dynamics model

Different molecular dynamics models have been developed to simulate the interaction of ultrafast XFEL pulses with matter. The first results have been pioneered by Neutze *et al.* [5]. In this model, the interaction of the free and quasi-free electrons with the atoms have been neglected. Bergh et al. [6] modeled the electrons as a continuous gas. Jurek et al. [7] included a treatment of the electrons as point-like particles, but neglected three-body-recombination that has a profound effect on the ionization dynamics in certain cases [8]. So far, the molecular dynamics models have been limited to only relatively small systems on the order of 10^3 to 10^4 atoms. Molecules to be imaged at the LCLS will have on the order of 10^4 to 10^6 atoms and more. Results obtained with a small number of atoms cannot simply be scaled to large molecules since electron capturing and the charge distribution inside of the molecule strongly depend on size, thereby affecting the Coulomb expansion dynamics.

Modeling soft-x-ray matter interaction

Since XFELs are not yet available, direct benchmarking of the simulation tools has not been possible. The models have to be modified in order to be applicable to x-ray matter interaction experiments performed at the FLASH facility. In particular, x-ray matter interaction is complicated by free-free (inverse Bremsstrahlung) absorption. In some models, this has been treated directly [9], whereas in other models [10] opacities have been calculated using a screened average ion model. In both cases it was found these models have been very successful in guiding and analyzing experiments performed at FLASH. Noticeable results include XFEL cluster interactions [9] and various coherent imaging experiments, including ultrafast coherent diffraction imaging [12], probing of FEL-induced sample explosion with fs time resolution [13], and 'disposable' multilayer optics [14].

SUMMARY AND CONCLUSIONS

XFEL light sources open up new areas of the laser-matter interaction. In this presentation, we have laid out the current understanding of the interaction of XFEL pulses with matter, and we have described some of the simulation approaches that are used to design experiments on future XFEL sources. Appropriately modified, these models have been successful in guiding and analyzing experiments performed at the extreme-ultraviolet FEL FLASH at wavelengths of 6 nm and longer. For several keV photon energies, no data from XFEL-matter interaction experiments is available yet but is anticipated to become available in the near future, which will allow to test our understanding in this wavelength regime.

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